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Numerical model of a long-term in situ diffusion and retention (DR) experiment in Opalinus Clay

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Abstract

Determining diffusion parameters under real conditions is necessary for the performance assessment of a deep geological repository. Here we present a numerical model for the interpretation of a long-term in situ diffusion and retention (DR) experiment performed on Opalinus clay (OPA) at Mont Terri underground rock laboratory in Switzerland using a comprehensive set of tracers which includes neutral (HTO, HDO), anionic (Cl, Br, I), weakly-sorbing (22Na, 133Ba and 85Sr) and strongly-sorbing tracers (60Co, 137Cs). Compared to the model used for scoping calculations, our current model accounts for the presence of a sintered filter, a gap between the filter and borehole wall and an excavation disturbed zone (EdZ). Sensitivity analyses have been performed to identify the relevance of these non-ideal effects and evaluate identifiability of diffusion anisotropy and distribution coefficients. Results of sensitivity analyses indicate that tracer concentrations are not sensitive to De of gap, but very sensitive to De of filter except for HTO and HDO. Concentrations of HTO, HDO, I, Na, Sr and Ba are sensitive to De of EdZ while those of strongly-sorbing tracers such as Co and Cs are much less sensitive. All sorbing tracers are sensitive to changes in Kd of clay. The relevance of diffusion anisotropy increases with time for all tracers except strongly-sorbing tracers.

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Key words: In situ diffusion, retention, numerical model, sensitivity analysis deuterium, tritium, bromide, iodide, sodium, strontium, cobalt, barium, caesium.

1. Introduction

Clay formations are considered potential host rocks for radioactive waste disposal in many countries. Clays have very small hydraulic conductivity and therefore molecular diffusion is the main radionuclide transport mechanism in these formations. Laboratory experiments are performed to improve our understanding of diffusion processes and determine key diffusion and retention parameters^{[1],[2]} (van Loon *et al.*, 2004; Samper *et al.*, 2006). *In situ* diffusion experiments are performed at underground research laboratories in clay formations to overcome the limitations of laboratory diffusion experiments and investigate possible scale effects. The *in situ* diffusion and retention (DR) experiment is one of such experiments being performed in Opalinus clay formation at Mont Terri underground rock laboratory in Switzerland. DR experiment is similar to previous *in situ* diffusion experiments such as DI-A^{[1],[3]} (van Loon *et al.*, 2004; Wersin *et al.*, 2004) and DI-B^[4] (Samper

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et al., 2006) (see Fig. 1). However, the concept has been optimized in order to determine diffusion anisotropy and evaluate diffusion and sorption parameters for strongly-sorbing tracers^[5] (Wersin *et al.*, 2007).

Scoping calculations for the design of DR were performed by Soler^[6] and Samper and Yang^{[2], [7]} (2006; 2007). Here we present a numerical model of DR experiment for evaluating the relevance of the presence of a sintered filter, a gap between the filter and borehole wall and an excavation disturbed zone (EdZ) and the identifiability of diffusion anisotropy and distribution coefficients.

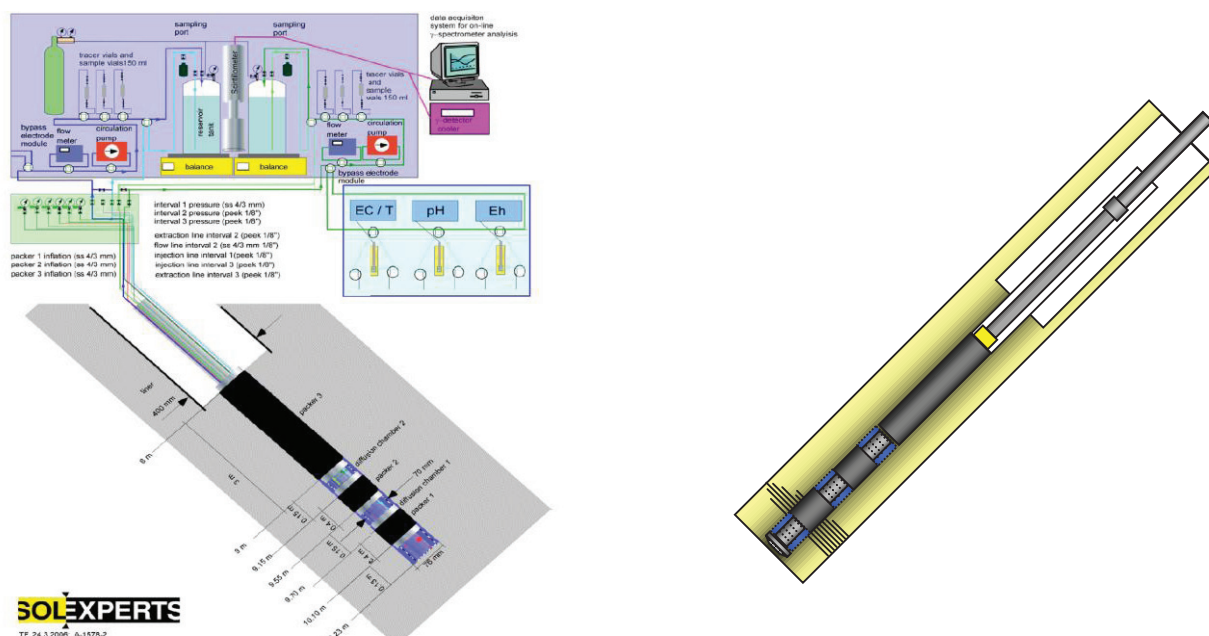


Fig. 1. Schematic drawing of the entire test setup (left) and borehole equipment for DR experiment (right). There are two tracer intervals and an auxiliary interval at the bottom for hydraulic observations (after SOLEXPERTS, Wersin *et al.*, 2007).

2. DR experiment

DR borehole was drilled at an angle of 45° with respect to the tunnel bottom so that it intersects the bedding at a right angle. DR includes several injection intervals which are shorter than those of previous diffusion experiments. That at the bottom serves as auxiliary interval for the observation of hydraulic pressure during the experiment. Tracer cocktails are injected into the upper intervals (Fig. 1). Each tracer interval is connected to the surface equipment with a tank of 20 L. Fluid circulates continuously to ensure tracers are well mixed at all times in the injection system where tracer concentrations are monitored in time. At the end of the experiment, the rock around the injection intervals will be overcored and tracer distribution profiles will be measured. Data on the time evolution of tracer concentrations in the injection system and tracer profiles in the rock will be used to derive effective diffusion coefficients, D_e , parallel and perpendicular to bedding, accessible porosity and sorption parameters. Borehole radius is 0.038 m. Tracer intervals are 0.15 m long and are separated 0.4 m. $^{60}\text{Co}^{2+}$, $^{137}\text{Cs}^+$, $^{133}\text{Ba}^{2+}$ and HDO are injected into the uppermost interval while Γ^- , Br^- , Cs^+ , $^{85}\text{Sr}^{2+}$, $^{22}\text{Na}^+$ and HTO are injected in the intermediate interval.

3. Numerical model

Opalinus clay exhibits diffusion anisotropy with effective diffusion parallel to bedding being approximately 4 times that perpendicular to bedding at Mont Terri laboratory. Dry density (ρ_d) and density of solids (ρ_s) of

Opalinus clay are equal to 2.33 and 2.74 g/cm³, respectively. Parameter values derived from laboratory through-diffusion experiments and other previous in situ diffusion experiments are listed in Table 1.

Given the symmetry with respect to borehole axis, numerical interpretation is performed with a 2-D axis-symmetric anisotropic model. Model domain is a 2-D vertical rectangle aligned with borehole axis (see Fig. 2). The left-hand-side boundary of the domain corresponds to borehole axis ($r = 0$). All other domain boundaries are no-flux boundaries. Fig. 2 shows the 2-D finite element grid. Model domain extends 1.5 m in radial direction and has a vertical thickness of 3 m. It is discretized with triangular finite elements by using COSMOS/M[®]. Elements are small near the borehole and increase far away (see Fig. 2). Numerical model considers five material zones: (1) Packed-off sections of borehole where tracers are injected; (2) Packer between the two injection intervals; (3) Gap; (4) Filter; and (5) Opalinus clay.

DR experiment is modelled using an approach similar to that used for modelling DI-B in situ diffusion experiment in Opalinus clay^[4] (Samper et al., 2006) and DIR in situ diffusion experiments in Callovo-Oxfordien clay^[7] (Samper et al., 2007). Numerical model has been used to: 1) Compute sensitivities of tracer concentrations to changes in model parameters^[8] (Samper and Yang, 2007); and 2) Interpret actual measured tracer data. Here we report mostly results of sensitivity analyses. Calculations are performed with CORE2D V4^[9] (Samper et al., 2003).

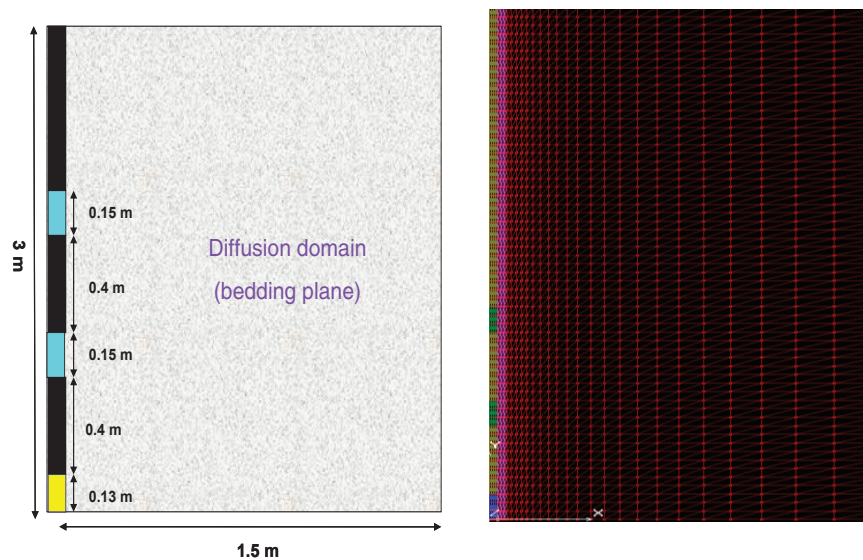


Fig. 2. Axial symmetric model domain of DR experiment (left) and 2-D finite element grid used for numerical mode (right).

Table 1. Transport parameters for scoping calculations of DR experiment Here D_p is pore diffusion coefficient, Φ_a is accessible porosity, $D_e = \Phi_a D_p$ is effective diffusion coefficient, α is rock capacity factor, K_d is distribution coefficient, $T_{1/2}$ is half-life, S is sorbed concentration, and C is solute concentration.

Tracer	Dp [m ² s ⁻¹]	De [m ² s ⁻¹]	Φa [m ³ m ⁻³]	α [m ³ m ⁻³]	Kd [m ³ kg ⁻¹]	T1/2 [a]	Remarks
HDO	3.3310-10	5. 10-11	0.15	0.15	0	stable	from DI-A1
HTO	3.3310-10	5. 10-11	0.15	0.15	0	12.323	from DI-A1
I ⁻	2. 10-10	1.6 10-11	0.08	0.08	0	stable	from DI-A1
²² Na ⁺	4.6710-10	7.10-11	0.15	0.62	2.04810-4	2.602	from DI-A1
⁸⁵ Sr ²⁺	4.6710-10	7. 10-11	0.15	3.13	1.3010-3	0.1777	preliminary from DI-A2
¹³³ Ba ²⁺	4.6710-10	7. 10-11	0.15	3.13	1.3 10-3	10.5	transport as Sr2+
¹³⁷ Cs ⁺	2. 10-9	3.10-10	0.15	1262.4	0.55	30.17	DI-A1, sorption at trace conc.
Cs ⁺	2 10-9	3.10-10	0.15	non linear: S =186·C ^{0.53}		stable	DI-A1, S [mg kg-1], C [mg L-1]
⁶⁰ Co ²⁺	2. 10-9	3.10-10	0.15	1262.4	0.55	5.272	transport as ¹³⁷ Cs ⁺

4. Sensitivity analyses

Samper and Yang^[7] (2007) performed sensitivity analyses to porosity, De and Kd. Here we complete such analysis by evaluating the relevance of the presence of a sintered filter, a gap between the filter and borehole wall and an excavation disturbed zone (EdZ) and the identifiability of diffusion anisotropy and distribution coefficients.

A series of sensitive runs have been performed by changing one-at-a-time effective diffusion of gap, filter and EdZ for all tracers and distribution coefficient of clay for sorbing tracers. Dimensions and parameters of gap, filter and EdZ are listed in Table 2. Effective diffusion coefficients for different materials were computed from those of intact clay by using Archie's law with an exponent of 1.33. Values of De are listed in Table 3.

Model results were computed for different combinations of presence or absence of gap, filter and EdZ. One can see in Fig. 3 that computed HTO activities are sensitive to EdZ and filter+gap. Results computed with gap, filter and EdZ fit better measured data than results obtained without gap, filter and EdZ. Similar conclusions are reached for other tracers (not shown here). Tracer concentrations are not sensitive to De of gap (not shown here). Except for HTO and HDO, tracer concentrations are clearly sensitive to De of filter (see Fig. 4). Concentrations of HTO, HDO, I, Na, Sr and Ba are sensitive to De of EdZ, while those of Co and Cs are not sensitive. This means that neutral, anionic and weakly-sorbing tracers are sensitive to De of EdZ, while strongly sorbing tracers are not sensitive to De of EdZ (not shown here). All sorbing tracers are sensitive to changes in Kd of clay (not shown here).

Anisotropy was analyzed by varying the ratio DeP (De parallel to bedding) to DeV (De normal to bedding) from 1 (isotropic) to 8. Model results indicate that diffusion anisotropy is not relevant for neutral, anionic and weakly-sorbing tracers for $t < 1$ year (Fig. 5), but as time increases anisotropy becomes more relevant. Anisotropy is not relevant at all for strongly-sorbing tracers such as Co and Cs.

5. Future work

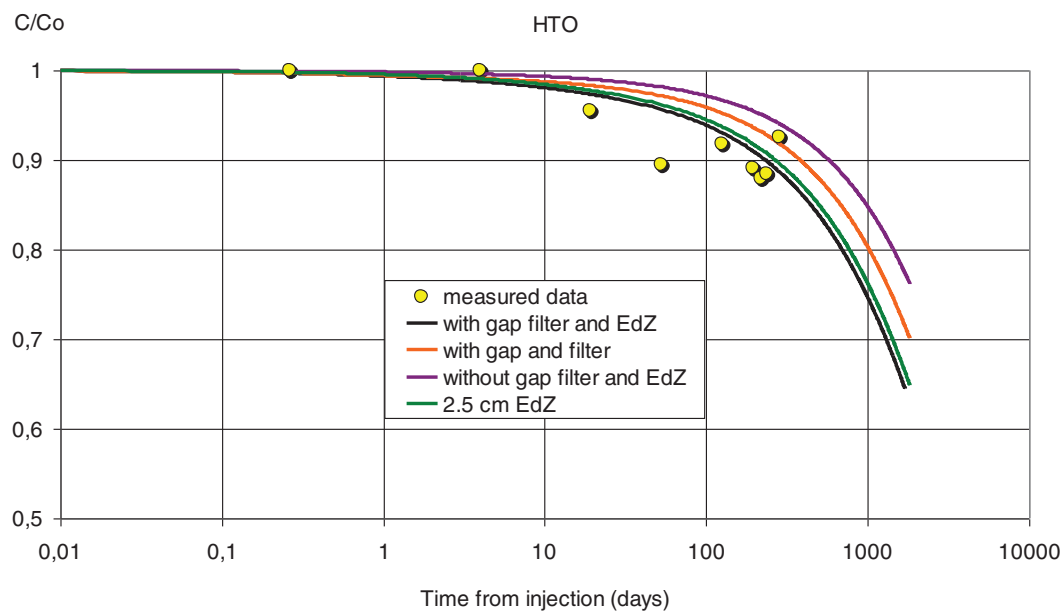
Future work will be devoted to: 1) Verify numerical solutions by code inter-comparison^[4] (Wersin et al., 2007) and discretization error analysis and 2) Check scoping calculations performed with a priori parameter values with measured data. Preliminary results indicate that there will be a need for parameter calibration for some tracers for which data are not now available such as Co and Ba (see Table 1). Such calibration should be done by solving the inverse problem using existing codes such as INVERSE-CORE^[10] (Dai & Samper, 2004). Such calibration should take into account sampling, measurement and analytical data errors and should rely on a wise choice of initial tracer activities, making sure that tracers have reached full mixing in tanks and circulation system.

Table 2. Dimensions and parameters for gap, filter and EdZ.

Material	Thickness (mm)	Φ
Inner gap	0.5	0.60
Filter	3	0.30
Gap between filter and clay	2	0.60
EdZ	20	0.30

Table 3. Effective diffusion coefficients of tracers for different materials computed from those in clay with Archie's law and an exponent of 1.33 (values in $10^{-11} \text{ m}^2/\text{s}$).

Materials/ Tracers	Gap	Filter	EdZ	Intact clay
HTO	31.7	6.27	12.6	5.0
$^{22}\text{Na}^+$	44.5	8.79	17.6	7.0
$^{85}\text{Sr}^{2+}$	44.5	8.79	17.6	7.0
I^-	10.1	2.00	4.02	1.6
^{134}Cs	190	37.6	75.5	3.0
Br^-	10.1	2.00	4.02	1.6
HDO	31.7	6.27	12.6	5.0
$^{60}\text{Co}^{2+}$	190	37.6	75.5	5.0
$^{133}\text{Ba}^{2+}$	44.5	8.79	17.6	7.0
$^{137}\text{Cs}^+$	190	3.76	75.5	5.0

**Fig. 3.** Sensitivity of computed HTO concentrations to the presence of gap, filter and EdZ (lines) and comparison to measured data (symbols).

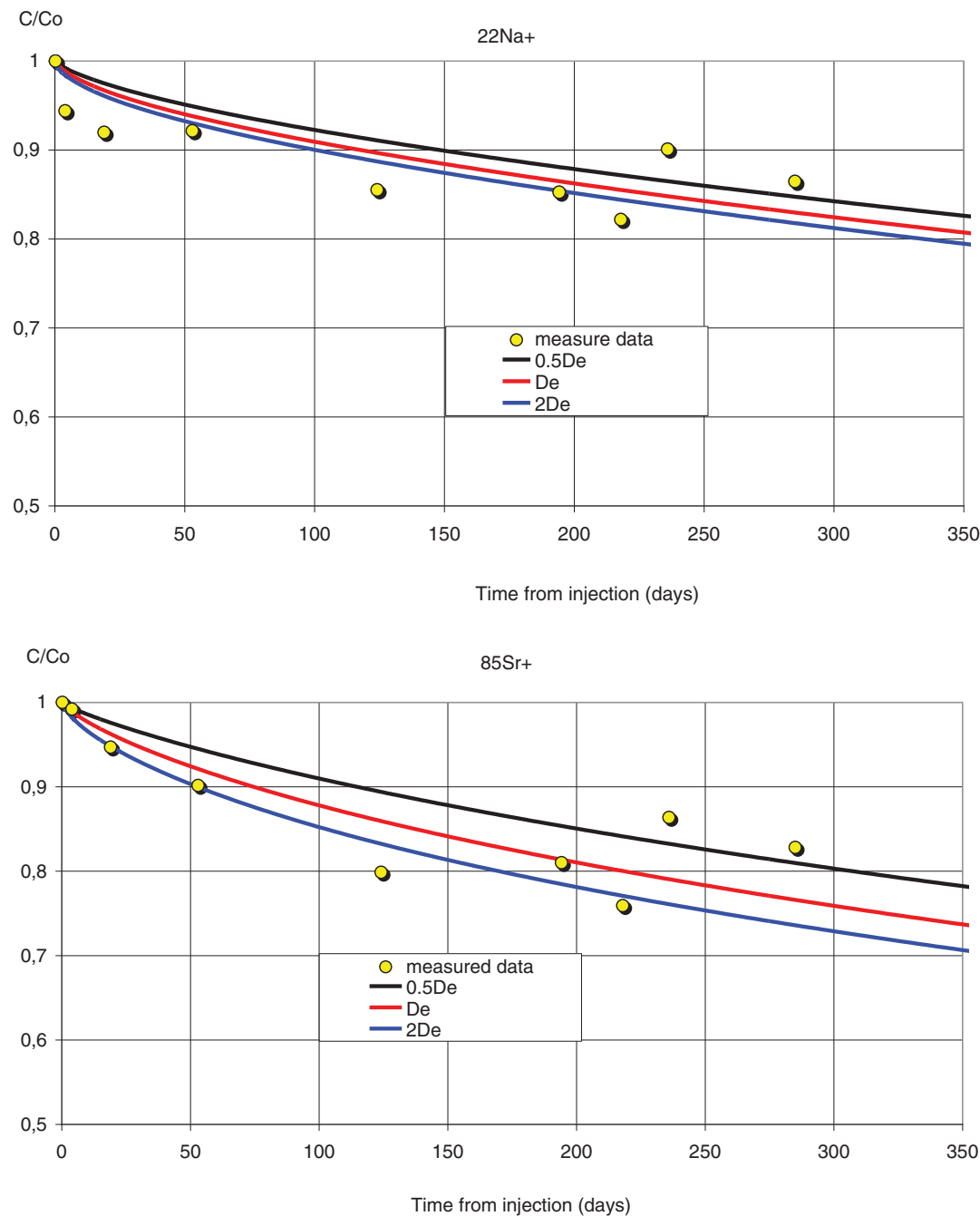


Fig. 4. Sensitivity of computed Na and Sr concentrations to changes in D_e of filter.

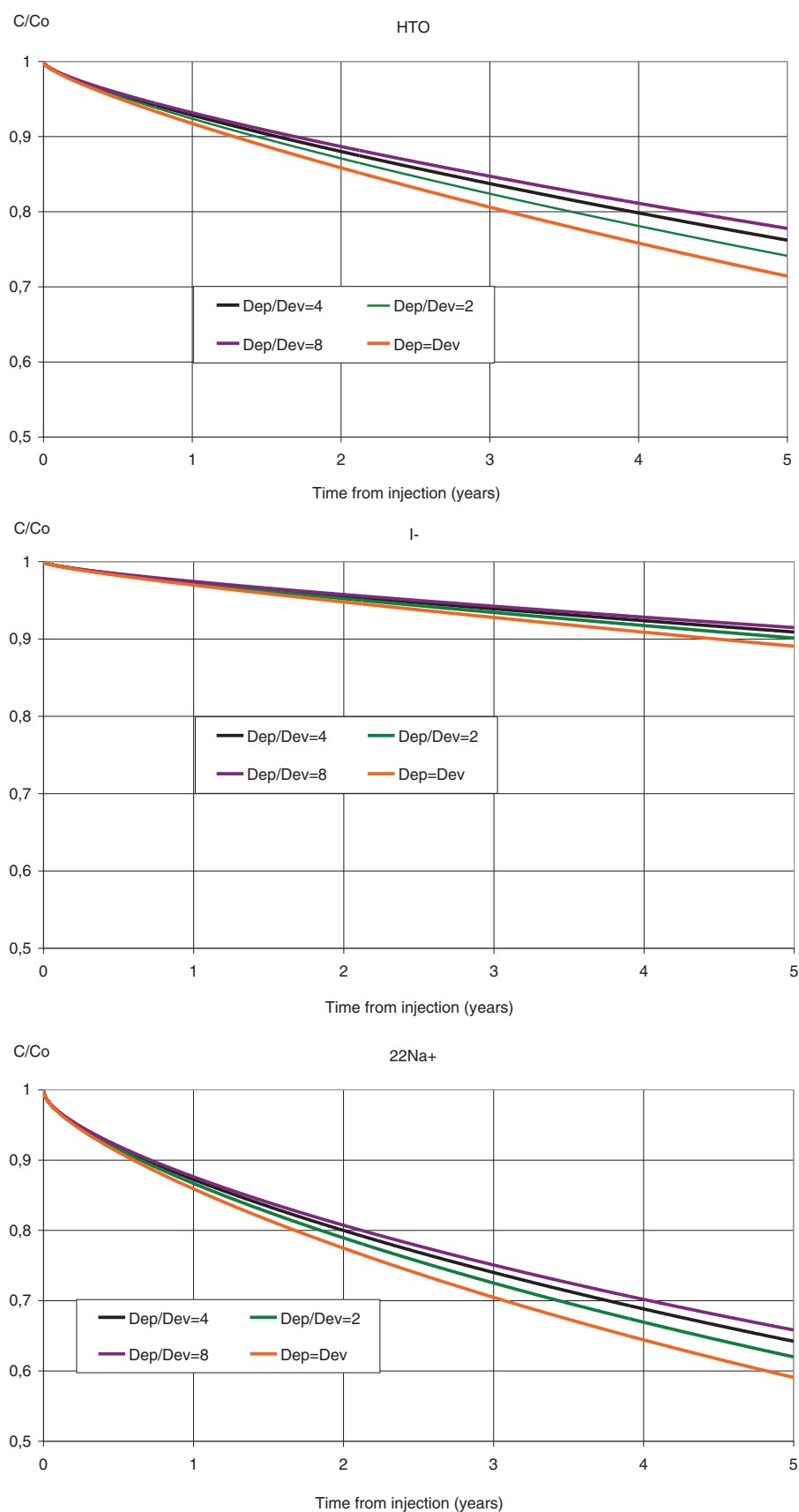


Fig. 5. Sensitivity of computed HTO (top), I⁻ (middle) and Na (bottom) concentrations to changes in the ratio D_{eP}/D_{eP} (D_{eP} parallel to bedding divided by D_{eP} normal to bedding).

6. Conclusions

Numerical models for the interpretation of DR experiment performed on Opalinus clay have been presented. Results of sensitivity analyses indicate that tracer concentrations are not sensitive to De of gap, but very sensitive to De of filter except for HTO and HDO. Concentrations of HTO, HDO, I, Na, Sr and Ba are sensitive to De of EdZ, while strongly-sorbing tracers such as Co and Cs are much less sensitive to that parameter. All sorbing tracers are sensitive to changes in Kd of clay. Relevance of diffusion anisotropy increases with time for all tracers except for strongly-sorbing tracers.

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